(19) Europäisches Patentamt
European Patent Office
Office européen des brevets

(11) EP 1 422 776 A1

(12) EUROPEAN PATENT APPLICATION
published in accordance with Art. 158(3) EPC

(43) Date of publication:
26.05.2004 Bulletin 2004/22

(21) Application number: **03746483.1**

(22) Date of filing: **14.04.2003**

(51) Int Cl.[7]: **H01M 8/02**, H01M 8/04, H01M 8/10, H01M 8/24

(86) International application number:
PCT/JP2003/004723

(87) International publication number:
WO 2003/088395 (23.10.2003 Gazette 2003/43)

(84) Designated Contracting States:
AT BE BG CH CY CZ DE DK EE ES FI FR GB GR HU IE IT LI LU MC NL PT RO SE SI SK TR
Designated Extension States:
AL LT LV MK

(30) Priority: **17.04.2002 JP 2002114552**

(71) Applicant: **MATSUSHITA ELECTRIC INDUSTRIAL CO., LTD.**
Kadoma-shi, Osaka 571-8501 (JP)

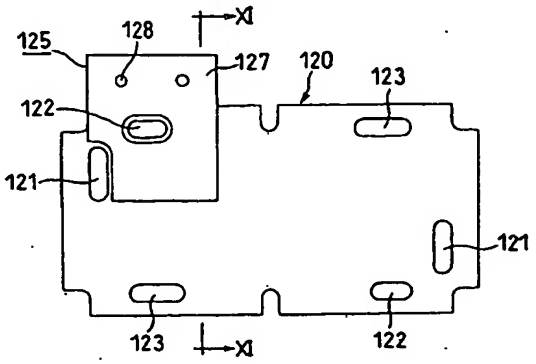
(72) Inventors:
- **HASE, Nobuhiro**
  Mino-shi, Osaka 562-0041 (JP)
- **HATOH, Kazuhito**
  Osaka-shi, Osaka 536-0015 (JP)
- **KUSAKABE, Hiroki**
  Sakai-shi, Osaka 590-0113 (JP)
- **OHARA, Hideo**
  Katano-shi, Osaka 576-0016 (JP)
- **KOBAYASHI, Susumu**
  Ikoma-shi, Nara 630-0134 (JP)
- **SHIBATA, Soichi**
  Hirakata-shi, Osaka 573-1149 (JP)
- **TAKEGUCHI, Shinsuke**
  Kadoma-shi, Osaka 571-0074 (JP)

(74) Representative: **Patentanwälte**
Leinweber & Zimmermann
Rosental 7,
II Aufgang
80331 München (DE)

(54) POLYMERIC ELECTROLYTE TYPE FUEL CELL

(57) A polymer electrolyte fuel cell comprises: a cell stack of a plurality of unit cells, each of the unit cells comprising a hydrogen-ion conductive polymer electrolyte membrane, an anode and a cathode sandwiching the polymer electrolyte membrane, an anode-side separator having a gas flow channel for supplying a fuel gas to the anode, and a cathode-side separator having a gas flow channel for supplying an oxidant gas to the cathode; a pair of current collector plates that sandwiches the cell stack; and a pair of end plates that clamps the cell stack and the current collector plates under pressure. The current collector plates comprise a conductive carbon material as a main component, and have a terminal section for connecting a power output cable in the vicinity of an inlet-side manifold for the fuel gas or the oxidant gas.

FIG. 10



Description

Technical Field

[0001] The present invention relates to a fuel cell for use in portable power sources, electric vehicle power sources, cogeneration systems or the like, particularly to a current collector plate of a polymer electrolyte fuel cell.

Background Art

[0002] A solid polymer electrolyte fuel cell generates electric power by electrochemically reacting a fuel gas containing hydrogen and an oxidant gas containing oxygen such as air through a polymer electrolyte membrane that selectively transports hydrogen ions.

[0003] The fuel cell generally comprises a cell stack of a plurality of unit cells, and each of the unit cells comprises an anode and a cathode sandwiching the polymer electrolyte membrane, an anode-side separator for supplying the fuel gas to the anode, and a cathode-side separator for supplying the oxidant gas to the cathode. The cell stack further includes a current collector plate for collecting current and an insulator plate for electrically insulating the cell stack from outside on each end of the cell stack and is sandwiched by end plates. The end plates are clamped by clamping means so that a clamping pressure of appropriate load is applied to the cell stack. The end plates are provided with means for supplying and discharging the fuel gas, oxidant gas and a coolant. A unit having such a structure is called a fuel cell stack.

[0004] In order to supply the cell stack with the fuel gas, oxidant gas and coolant, at least one of the current collector plates positioned at both ends has through holes through which the gases or coolant flows. In order to prevent the corrosion of these through holes, and to reduce the contact resistance to the separator of the neighboring cell and improve the electric conductivity of the current collector plate itself in the plane direction for suppressing the loss due to the electric resistance, the current collector plate is commonly a metallic material, such as stainless steel or copper, plated with gold. The gold-plated current collector plate will corrode from minutes pits if the thickness of the gold plating is insufficient. Thus, an anti-corrosion structure that eliminates the direct contact of the gases has been proposed for the through holes for supplying and discharging the gases and coolant.

[0005] However, the use of the gold-plated metallic current collector plate presents the following problems. In order that the polymer electrolyte fuel cell functions normally, the water content of the polymer electrolyte membrane must be high, so the supplied gases and the discharged gases contain large amounts of steam. Therefore, the metallic current collector plate is liable to corrosion at its parts contacting the supplied gases and the gases to be discharged. In case of the corrosion of metal at the gas supply route, metal ions generated thereby are included, impairing the performance of the electrolyte membrane. Also, in case of the corrosion of metal at the coolant supply route, the insulating property of the coolant is deteriorated to cause a leak current, resulting in a decrease in the amount of power generation.

[0006] In order to prevent the inclusion of ions due to the corrosion of metal, it is necessary to either increase the thickness of the gold-plating or employ an anti-corrosion structure. However, increasing the thickness of the gold-plating is very costly. Also, employing an anti-corrosion structure involves an increase in the number of components and production steps, so that the cell structure and assembling operation of the components become complicated. There is also an attempt to use a carbon material, which is free from the fear of ion leaching due to corrosion, for collecting current. The carbon material, however, is brittle and has higher electric resistivity than metal, thereby necessitating the current collector plate to have sufficient thickness.

Disclosure of Invention

[0007] The present invention provides a polymer electrolyte fuel cell having improved current collector plates. This fuel cell comprises: a cell stack of a plurality of unit cells, each of the unit cells comprising a hydrogen-ion conductive polymer electrolyte membrane, an anode and a cathode sandwiching the polymer electrolyte membrane, an anode-side separator having a gas flow channel for supplying a fuel gas to the anode, and a cathode-side separator having a gas flow channel for supplying an oxidant gas to the cathode; a pair of current collector plates that sandwiches the cell stack; and a pair of end plates that clamps the cell stack and the current collector plates under pressure. The current collector plates in accordance with the present invention comprise a conductive carbon material as a main component, and have a terminal section for connecting a power output cable in the vicinity of an inlet-side manifold for the fuel gas or the oxidant gas.

[0008] The terminal section preferably has a coating layer comprising a good electric conductor.

[0009] The coating layer comprising a good electric conductor is more preferably provided in a region that extends from the terminal section to a part corresponding to an inlet-side part of the gas flow channel of the cell communicating with the inlet-side manifold.

Brief Description of Drawings

[0010]

FIG. 1 is a partially sectional side view of an example of the structure of a fuel cell stack in accordance with the present invention.

FIG. 2 is a front view of a current collector plate in Embodiment 1 of the present invention.
FIG. 3 is a back view of the same current collector plate.
FIG. 4 is a sectional view taken on line IV-IV of FIG. 2.
FIG. 5 is a perspective view of the main part of another example of connecting a cable to the current collector plate.
FIG. 6 is a front view of a current collector plate in Embodiment 2 of the present invention.
FIG. 7 is a back view of another example of the current collector plate.
FIG. 8 is a front view of an example of an insulator plate to be used with the current collector plate.
FIG. 9 is a front view of an anode-side separator in Embodiment 3 of the present invention.
FIG. 10 is a front view of a current collector plate to be combined with the separator plate of FIG. 9.
FIG. 11 is a sectional view taken on line XI-XI of FIG. 10.
FIG. 12 is a front view of a cathode-side separator in Embodiment 3 of the present invention.
FIG. 13 is a front view of a current collector plate to be combined with the separator plate of FIG. 12.
FIG. 14 is a sectional view taken on line XIV-XIV of FIG. 13.
FIG. 15 is a front view of another anode-side separator.
FIG. 16 is a front view of a current collector plate to be combined with the separator of FIG. 15.
FIG. 17 is a front view of another cathode-side separator.
FIG. 18 is a front view of a current collector plate to be combined with the separator of FIG. 17.
FIG. 19 is a longitudinal sectional view of another example of the current collector plate.
FIG. 20 is a front view of an anode-side current collector plate in Embodiment 4 of the present invention.
FIG. 21 is a front view of a cathode-side current collector plate in Embodiment 4 of the present invention.
FIG. 22 is a front view of an anode-side current collector plate in Embodiment 5 of the present invention.
FIG. 23 is a back view of the anode-side current collector plate of FIG. 22.
FIG. 24 is a front view of an anode-side current collector plate in Embodiment 6 of the present invention.
FIG. 25 is a sectional view taken on line XXV-XXV of FIG. 24.
FIG. 26 is an enlarged sectional view of the main part of an example of a good electric conductor layer formed on the surface of a core metal plate.

Best Mode for Carrying Out the Invention

[0011] The inventors of the present invention have examined the use of a plate or a molded plate mainly composed of a carbon material that is free from the fear of corrosion as the current collector plate. As a result, they have found that when the current collector plates have a current-output terminal section in the vicinity of an inlet-side manifold for the reaction gas, particularly the fuel gas, effects unique to a polymer electrolyte fuel cell can be obtained. That is, the electrodes are humidified by water generated by the reaction on the inlet side of the reaction gas supplied to the electrodes. Further, the polymer electrolyte membranes which exhibit hydrogen-ion conductivity when moistened with water can be effectively humidified.

[0012] In order to improve the durability of the electrodes, it is said to be effective to highly humidify the electrodes for operation. To highly humidify the electrodes, an attempt to supply highly humidified reaction gases to the cell is being made. However, an increase in the degree of humidification of the fuel gas leads to a decrease in energy efficiency. According to the present invention, the terminal section is provided in the vicinity of an inlet-side reaction gas manifold of the current collector plates. Thus, the current density at the electrode-reaction parts adjacent to the inlet-side manifold is heightened and the amount of water generated by the reaction is therefore increased, so that the electrodes can be kept in a highly humidified condition.

[0013] A current collector plate comprising a conductive graphite plate or a molded plate of a composite material of a conductive carbon material and a binder has larger electric resistivity than a metallic current collector plate even if it is made electrically anisotropic such that the electric resistivity in the plane direction is smaller as described later. Thus, in the plane of the current collector plate, a part close to the terminal section has smaller electric resistance to the terminal section than a distant part. Accordingly, there is a difference in current density between the part close to the terminal section and the distant part, and the electrode reaction increases at the former part having higher current density. In order to further enhance the current density on the inlet side of the reaction gas at the electrode, it is preferable to provide a coating layer comprising a good electric conductor in a region extending from the terminal section of the current collector plate to the part corresponding to the inlet-side part of the reaction gas of the electrode.

[0014] When the current density on the inlet side of the fuel gas at the anode is heightened as described above, the amount of water generated by the reaction at the corresponding cathode is increased, and the generated water moves to the anode side by reverse diffusion. In this way, by highly humidifying the inlet part of the reaction gas which is not expected to be humidified by the water generated at the electrode section, particularly the vicinity of the inlet part of the fuel gas, elec-

trode durability can be improved. Also, the degree of humidification of the fuel gas supplied to the cell can be decreased, and energy efficiency can be improved.

[0015] In another preferable mode of the present invention, the current-output terminal section of the current collector plate has a coating layer comprising a metal film or plate, which is a good electric conductor. This makes it possible to reduce the loss due to the electric resistance at the terminal section resulting from the use of carbon having lower conductivity than metal and to improve the strength of the terminal section. In a more preferable mode, the metal plate constituting the coating layer is extended so as to overlap with the separator, i. e., the metal plate is applied with the clamping pressure of the cell stack. Thus, the reinforcing effect can be further improved.

[0016] Also, the inventors of the present invention have examined the use of such a current collector plate that the electric resistivity in the plane direction is smaller than the electric resistivity in the thickness direction, i.e., the stacking direction of the stack. As a result, by compression molding a composite material of a highly graphitized carbon material and a binder, they have succeeded in obtaining an electrically anisotropic current collector plate In which the electric resistivity in the plane direction is smaller. This current collector plate is thin and capable of preventing performance deterioration caused by metal ions.

[0017] In still another preferable mode of the present invention, the current collector plate is integrated with the adjoining separator. This can reduce the number of components as well as the loss due to the electric resistance caused by contact resistance.

[0018] In another preferable mode of the present invention, a metal plate for connecting a power output cable is clamped to the terminal section of the current collector plate at two or more points. This can prevent the cracking of the terminal section of the current collector plate.

[0019] A suitable example of the current collector plate comprising a conductive carbon as a main component which is used in the present invention is a conductive carbon plate, such as a commercially available glassy carbon plate or an expanded graphite plate, which is worked into a predetermined shape by cutting, abrasive blasting or the like. The conductive carbon plate preferably has an electric resistivity of approximately 1 mΩ·cm or less in the plane direction. A more preferable current collector plate is made of a molding material prepared by adding a graphite powder to a thermoplastic resin, such as polyphenylene sulfide or polypropylene, or a thermoplastic resin, such as epoxy resin or phenol resin, or a mixture thereof, and is obtained by forming the molding material into a predetermined shape by compression molding, injection molding, or the like. It is also possible to use a plate obtained by forming such a molding material in a plate and working the plate into a predetermined shape by cutting or abrasive blasting. These molded plates preferably have an electric resistivity of approximately 5 mΩ·cm or less in the plane direction.

[0020] In the case of molding the separators of a fuel cell, it is desirable to mold the current collector plates out of the same molding material in terms of material cost reduction. With regard to the thickness of the current collector plate, the terminal section and the other part may have a different thickness. In changing the thickness, it is desirable to employ such a structure that the difference in thickness is eased by a curved surface in order to prevent the occurrence of breaking of the material caused by the concentration of stress on the base portion of the terminal section. Also, the thickness of the current collector plate needs to be at least about 3 to 6 mm so that it will not break.

[0021] The present invention is more specifically described below by way of embodiments.

Embodiment 1

[0022] FIG. 1 shows an example of a polymer electrolyte fuel cell stack in this embodiment.

[0023] An electrolyte membrane electrode assembly (MEA) 1 consists of a polymer electrolyte membrane and an anode and a cathode sandwiching the polymer electrolyte membrane. The polymer electrolyte membrane has a size slightly larger than the electrodes. The periphery of the polymer electrolyte membrane is sandwiched by gaskets. This structure of the MEA is widely known. The MEAs 1 and separators are alternately stacked. Two kinds of separators are used; a single separator 2 has an oxidant gas flow channel on one side and a fuel gas flow channel on the other side and serves both as a cathode-side separator and an anode-side separator, and a composite separator 2A has a coolant flow channel that is formed between a cathode-side separator and an anode-side separator.

[0024] An end plate 5a is joined to the upper end of the above-described cell stack with a cathode-side current collector plate 3a and an insulator plate 4a, while an end plate 5b is joined to the lower end of the stack with an anode-side current collector plate 3b and an insulator plate 4b. The end plates 5a and 5b are clamped by bolts 6 and nuts 8, so that a predetermined load is applied to the cell stack. 7 represents a spring. The end plate 5a has inlets/outlets for the reaction gases and coolant. In this figure, an oxidant gas outlet 11, a fuel gas inlet 12 and a coolant outlet 13 are illustrated. In this way, a polymer electrolyte fuel cell stack 10 is assembled.

[0025] The current collector plates 3a and 3b have terminal sections 15a and 15b, respectively, which protrude from the side of the cell stack, and power output cables 16a and 16b are connected to these terminal sections by metal fittings 15a and 15b, respectively. In FIG. 1, the current collector plates 3a and 3b are illustrated as serving also as a cathode-side separator and

an anode-side separator, respectively, but this is not to be construed as limiting the current collector plates as specifically described below.

[0026] FIGs. 2 to 4 illustrate a current collector plate serving also as a cathode-side separator. FIG. 2 is a front view of the cathode side of the current collector plate, FIG. 3 is a back view thereof, and FIG. 4 is a sectional view taken on line IV-IV of FIG. 2. In FIG. 3, the power output cable, the metal plates for connecting the cable, etc., are omitted.

[0027] A current collector plate 30 has a pair of oxidant gas manifold apertures 31, a pair of fuel gas manifold apertures 32, and a pair of coolant manifold apertures 33. The current collector plate 30 has an oxidant gas flow channel 34 that communicates with the pair of manifold apertures 31 on the cathode facing side. On the insulator-plate-facing side, the current collector plate 30 has grooves 37, 38 and 39 surrounding the manifold apertures 31, 32 and 33, respectively, and O-rings are fitted into these grooves. The O-rings fitted into these grooves are compressed between the current collector plate and the insulator plate to prevent leakage of the gases and coolant from the respective manifold apertures.

[0028] The current collector plate 30 further has a terminal section 35 with two holes 36, and two metal plates 45 are fastened to the terminal section so as sandwich it with screws 46 and nuts 47. 48 represents a spring washer, while 49 represents a flat washer. A core wire 41 of a power output cable 40 is connected to the metal plates 45 with solder 42. In FIG. 2, of the pair of oxidant gas manifold apertures 31 and the pair of fuel gas manifold apertures 32, the one closer to the terminal section 35 is the inlet-side manifold aperture.

[0029] With regard to the connecting points between the terminal section 35 of the current collector plate 30 and the metal plates 45 for connecting the power output cable, there is no particular limitation, but the connecting points are preferably two or more as illustrated in FIGs. 2 and 3. The reason is that this equalizes the pressure of the contact surface, thereby lessening the loss caused by the resistance and leveling out the stress produced by clamping. Also, in addition to connecting the metal plates to both sides of the terminal section as described above, the metal plate may be connected to only one side of the terminal section. However, connecting the metal plates to both sides produces the effect of increasing the contact surface and reducing the contact resistance. Further, this equalizes the stress applied to the terminal section and therefore produces the effect of preventing the breaking caused by excessive clamping.

[0030] FIG. 5 shows another example of connecting the power output cable 40 to the terminal section 35 of the current collector plate 30. The terminal section 35 of the current collector plate 30 is fitted into a U-shaped metal fitting 43 to which the cable 40 is soldered, and they are fastened together with a screw 44 going through the hole 36.

[0031] In this embodiment, the fuel cell stack was clamped using clamping means consisting of bolts, springs and nuts, but other clamping means may be employed. The power output cable was connected to the terminal section of the current collector plate by making holes in the terminal section and connecting the metal plates thereto with the screws and nuts, but it may be connected in other methods. For example, it may be connected by making a screw hole in the terminal section 35 and screwing a pressure terminal connected to the power output cable in the hole. Another connecting method capable of stable electrical connection may also be employed.

[0032] Also, the power output cable and the metal plates for connecting the cable were connected with solder, but they may be connected by any methods capable of good electrical connection, e.g., joining through pressure by caulking. As the material of the metal plates for connecting the cable, a metallic material having low contact resistance may be used. For example, metal having low electric resistance and good workability, such as phosphor bronze or copper, may be used.

Embodiment 2

[0033] A method for reducing the resistance loss at the terminal section of the current collector plate is described.

[0034] FIG. 6 is a front view of the cathode side of a current collector plate in this embodiment. A current collector plate 30A has a metal film 50 of about 1 μm in thickness formed on each side of a terminal section 35. Except for this, it has the same structure as the current collector plate 30 of FIG. 2. The metal film 50 makes it possible to reduce the contact resistance, for example, between the terminal section 35 of the current collector plate and the metal plate for connecting the power output cable.

[0035] FIG. 7 shows still another example of the current collector plate. A current collector plate 30B has a metal film 51 of about several μm to tens of μm in thickness on the substantially whole surface of the insulator-plate-facing side. The metal film makes it possible to improve the conductivity in the plane direction of the current collector plate and further reduce the resistance loss. In this case, in order to prevent the performance deterioration caused by the leaching of metal ions, it is desirable not to form the metal film at the locations contacting the oxidant gas, fuel gas and coolant. In this case, the metal film is not formed around grooves 37, 38 and 39 which surround the respective manifold apertures and to which O-rings are fitted. As the metal forming the metal film, a conductive material such as copper and aluminum may be used. The film may be formed, for example, by vapor deposition or thermal spraying, and thermal spraying is preferable because it can form a film of a predetermined thickness in a short

period of time. Except for this, the current collector plate 30B has the same structure as that of Embodiment 1.

[0036] FIG. 8 illustrates an insulator plate to be disposed on the backside of the current collector plate of FIG. 2 or FIG. 6. An insulator plate 60 has a pair of oxidant gas manifold apertures 61, a pair of fuel gas manifold apertures 62, and a pair of coolant manifold apertures 63.

Embodiment 3

[0037] This embodiment describes a current collector plate independent of a separator.

[0038] FIG. 9 is a front view of an anode-side separator, and FIG. 10 is a front view of a current collector plate in contact with the anode-side separator. FIG. 12 is a front view of a cathode-side separator, and FIG. 13 is a front view of a current collector plate in contact with the cathode-side separator.

[0039] An anode-side separator 110 has a pair of oxidant gas manifold apertures 111, a pair of fuel gas manifold apertures 112, and a pair of coolant manifold apertures 113, and further has a fuel gas flow channel 116 communicating with the pair of manifold apertures 112 on the anode facing side. A current collector plate 120 to be disposed on the backside of the separator 110 has a pair of oxidant gas manifold apertures 121, a pair of fuel gas manifold apertures 122, and a pair of coolant manifold apertures 123. The current collector plate 120 has a terminal section 125 in the vicinity of the inlet-side manifold aperture (the upper left manifold aperture in the figure, as is clear from the arrows showing the direction of the gas flow) of the pair of fuel gas manifold apertures of the separator 110, the terminal section 125 protruding outward. A coating layer 127 comprising a good electric conductor is provided at the terminal section 125 and its adjacent part, i.e., a region 126 corresponding to the inlet-side manifold aperture of the separator 120 and the inlet-side part of the gas flow channel communicating therewith. The region 126 is thinner than the other part such that it becomes as thick as the other part when the coating layer 127 is formed on each side of the region 126. The terminal section 125 has holes 128 for mounting the metal plates for connecting the power output cable.

[0040] A cathode-side separator 130 has a pair of oxidant gas manifold apertures 131, a pair of fuel gas manifold apertures 132, and a pair of coolant manifold apertures 133, and further has an oxidant gas flow channel 134 communicating with the pair of manifold apertures 131 on the cathode facing side. A current collector plate 140 to be disposed on the backside of the separator 130 has a pair of oxidant gas manifold apertures 141, a pair of fuel gas manifold apertures 142, and a pair of coolant manifold apertures 143. The current collector plate 140 has a terminal section 145 in the vicinity of the inlet-side manifold aperture (the upper right manifold aperture in the figure) of the pair of oxidant gas manifold apertures 131 of the separator 130, the terminal section 145 protruding outward. A coating layer 147 comprising a good electric conductor is provided at the terminal section 145 and its adjacent part, i.e., a region 146 corresponding to the inlet-side manifold aperture of the separator 130 and the inlet-side part of the gas flow channel communicating therewith. The region 146 is thinner than the other part such that it becomes as thick as the other part when the coating layer 147 is formed on each side of the region 146. The terminal section 145 has holes 148 for mounting the metal plates for connecting the power output cable.

[0041] As is clear from FIGs. 9 and 10, the coating layer 127 of the terminal section 125 of the current collector plate 120 extends to the part corresponding to the inlet-side part of the gas flow channel 116 of the separator plate 110. The coating layer 127 comprising a metal film or a metal plate has smaller electric resistance than the other part composed mainly of the carbon material. Thus, when current is output from the terminal section 125 during power generation, the current density is higher at the above-described part. Thus, the electrode reaction of the cell proceeds more actively on the inlet side of the gas flow channel 116. Likewise, on the cathode side, it proceeds more actively on the inlet side of the gas flow channel 134. In this way, in the respective cells, the amount of water generated by the reaction becomes increased in the vicinity of the inlet-side gas manifold aperture, so that the reaction gases supplied to its downstream are humidified.

[0042] As illustrated in FIGs. 10 and 13, it is preferable that the terminal section 125 of the anode-side current collector plate and the terminal section 145 of the cathode-side current collector plate face each other, i.e., the inlet-side fuel gas manifold aperture and the inlet-side oxidant gas manifold aperture be in close vicinity to each other.

[0043] FIGs. 15 to 18 show modified examples of the combination of the separator and the current collector plate.

[0044] An anode-side separator 110A of FIG. 15 is the same as the separator 110 of FIG. 9 except that the position of oxidant manifold apertures 111A is changed. A current collector plate 120A to be disposed on the backside of the separator 110A is the same as the current collector plate 120 of FIG. 10 except that the position of oxidant manifold apertures 121A is changed and that the shape of coating layers 127A is slightly different.

[0045] Also, a cathode-side separator 130A of FIG. 17 is the same as the separator 130 of FIG. 12 except that the position of oxidant manifold apertures 131A is changed. A current collector plate 140A to be disposed on the backside of the separator 130A is the same as the current collector plate 140 of FIG. 13 except that the position of oxidant manifold apertures 141A is changed and that the shape of coating layers 147A is slightly different.

[0046] Each of the coating layers 127, 127A, 147 and

147A may comprise the metal film as described in Embodiment 2. However, in a more preferable mode, the coating layer may comprise a metal plate.

[0047] It is preferable that part of the coating layer comprising a metal plate be positioned inside the projected area of the separator stacked on the current collector plate, as illustrated in FIG. 9. In this case, the clamping load of the stack is applied to the metal plate via the separator and the current collector plate. Then, the metal plates are firmly secured depending on the clamping load of the stack. Since the metal plates also cover the terminal section of the current collector plate, they can serve to effectively alleviate the stress applied to the base portion of the terminal section by the cable connected to the terminal section.

[0048] It is preferable that the metal plates be bonded to the current collector plate with an adhesive having elasticity such as a conductive adhesive or a silicone adhesive. Insulating adhesives such as the silicone adhesive are convenient for restricting the current density of a specific part of the current collector plate by interposing it between the current collector plate and the metal plate.

[0049] FIG. 19 shows an example of bonding the metal plates 127 to the region 126 of the current collector plate 120 with a conductive adhesive 129. As illustrated therein, when the metal plates are fixed to the current collector plate with the adhesive such that there are no gaps between the current collector plate and the metal plates, the stress applied to the terminal section is effectively alleviated. When the metal plates are bonded to the current collector plate with the conductive adhesive, it has been confirmed that the voltage between the current collector plates at both ends of the stack is higher than that without the adhesive by about 5 mV at the time of an output of a current of 30 A by the terminal sections of the current collector plates.

Embodiment 4

[0050] FIGs. 20 and 21 illustrate current collector plates of this embodiment.

[0051] FIG. 20 is a front view of an anode-side current collector plate. A current collector plate 120B has the same structure as the current collector plate of FIG. 10 except that it does not have the coating layer 127 at a terminal section 125B and its adjoining part, and is disposed on the backside of the anode-side separator 110 of FIG. 9.

[0052] FIG. 21 is a front view of a cathode-side current collector plate. A current collector plate 140B has the same structure as the current collector plate of FIG. 13 except that it does not have the coating layer 147 at a terminal section 145B and its adjoining part, and is disposed on the backside of the cathode-side separator 130 of FIG. 12.

[0053] Each of these current collector plates comprises a molded plate of a composite material of a carbon material and a binder. Therefore, during power generation, the current density becomes higher in the vicinity of the terminal section, i.e., in the vicinity of the inlet-side gas manifold aperture, which produces the effect of humidifying the gases in the same manner as in Embodiment 4.

Embodiment 5

[0054] This embodiment explains an example of a current collector plate serving also as a separator.

[0055] FIG. 22 is a front view of a current collector plate serving also as an anode-side separator, and FIG. 23 is a back view thereof. A current collector plate 150 has a pair of oxidant gas manifold apertures 151, a pair of fuel gas manifold apertures 152, and a pair of coolant manifold apertures 153, and has a fuel gas flow channel 156 communicating with the pair of manifold apertures 152 on the anode facing side. The current collector plate 150 has a terminal section 155 in the vicinity of the inlet-side manifold aperture (the upper left manifold aperture in FIG. 22) of the pair of anode-side manifold apertures 152, and it has a coating layer 157 extending to the vicinity of the nearby manifold aperture 151 on the backside. This coating layer has the same structure, for example, as the coating layer of the current collector plate 120 as described in Embodiment 3 except it is formed only on the backside of the current collector plate. Although this embodiment has described the anode-side current collector plate, it will be apparent to those skilled in the art that a current collector plate serving as a cathode-side separator can also be formed in the same manner.

Embodiment 6

[0056] This embodiment describes a current collector plate into which a metal plate serving as a core member is buried.

[0057] FIG. 24 is a front view of a current collector plate of this embodiment, and FIG. 25 is a sectional view taken on line XXV-XXV of FIG. 24. A current collector plate 160 is produced by molding a molding material of a conductive carbon material and a binder so as to surround a metal plate serving as a core member. The current collector plate 160 has a pair of oxidant gas manifold apertures 161, a pair of fuel gas manifold apertures 162, and a pair of coolant manifold apertures 163, and further has a terminal section 165 protruding sideward. A metal plate 167 buried in this current collector plate has a smaller external size than the current collector plate such that it is not exposed outside, and it has larger apertures than the manifold apertures at its corresponding parts.

[0058] FIG. 26 shows an example of forming a good electric conductor layer 169 on the surface of the core metal plate. This layer makes it possible to reduce the contact resistance between the layer comprising the

molding material and the core metal plate and improve the electric characteristics of the current collector plate. The good electric conductor layer can be formed by removing the oxides on the surface of the core metal plate and forming a layer of noble metal, conductive inorganic oxide, conductive inorganic nitride, or conductive inorganic carbide on the core metal plate.

[0059] The following describes an example of molding a current collector plate, using a molding conductive material of a mixture of 80 wt% graphite and 20 wt% phenol resin, in which a core metal plate is buried.

[0060] First, 50 g of a molding compound was evenly charged into a mold and was compressed preliminarily at a mold clamping pressure of 100 kgf/cm^2. At this time, the temperature of the mold was 70°C. Next, the mold was opened, and a metal plate was inserted into the mold. Then, 50 g of the molding compound was charged therein, the mold was clamped at a mold clamping pressure of 500 kgf/cm^2, and the temperature was raised to 160°C. The resultant current collector plate had greater strength and lower electric resistance than the current collector plate molded only of the conductive molding material (7 mm in thickness), and hence it became possible to reduce the thickness of the current collector plate to 4 mm. As the core member, a brass plate of 2 mm in thickness was used. The current collector plate had a pair of manifold apertures for each of the oxidant gas, fuel gas and coolant, as illustrated in FIG. 24. Since the inner faces forming the manifold apertures were covered with the conductive molding material, the core metal plate was prevented from coming in contact with the fluids flowing through the respective manifold apertures. A stack comprising these current collector plates was assembled, and using pure water as the coolant, a power generation test was performed for 100 hours. The number of the stacked cells was 50. As a comparative example, a stack of 50 cells comprising current collector plates made of only brass was also tested in the same manner. As a result, corrosion was observed on the inner faces of the coolant manifold apertures formed in the brass current collector plates of the comparative example, but corrosion was not observed on the inner faces of the manifold apertures in the current collector plates of this example. The pure water used as the coolant was analyzed after the test. As a result, from the cooling water of the stack comprising the brass current collector plates, copper ions of 100 ppm and zinc ions of 80 ppm were detected. In contrast, from the coolant of the stack of this example, no metal ions were detected. By covering a metal core member with a conductive molding material, it becomes possible to ensure the strength and electric conductivity and reduce the cost and the leaching of impurities.

[0061] Examples of forming a good electric conductor layer on the surface of a core metal plate are specifically described below.

(1) A layer of platinum or gold having a thickness of 120 A was formed on the surface of a brass plate having a thickness of 1 mm by a sputtering method using an RF-planar magnetron. As the target, platinum (99%) or gold (99%) was used, and the substrate temperature was 500°C. The sputtering atmosphere was Ar (99.9999%) of 4×10^{-2} Torr, the sputtering power was 400 W, and the deposition rate was 1.5 μm/hr.

(2) A TiN layer having a thickness of 1 μm was formed on the surface of a Ti plate having a thickness of 1 mm by a sputtering method using an RF-planar magnetron. As the target, TiN (99%) was used, and the substrate temperature was 500°C. The sputtering atmosphere was Ar (99.9999%) of 4×10^{-2} Torr, the sputtering power was 400 W, and the deposition rate was 1.5 μm/hr. The resultant sputtered layer was identified as TiN by structural analysis of X-ray diffraction. The resistivity of the TiN layer obtained by this method was 2×10^{-4} Ω · cm.

(3) A Ti-Al-N layer having a thickness of 1.2 μm was formed on the surface of an Al plate having a thickness of 1 mm by a sputtering method using an RF-diode. As the target, Ti-Al-N (99%) was used, and the substrate temperature was 300°C. The sputtering atmosphere was Ar (99.999%) of 4×10^{-2} Torr, the sputtering power was 300 W, and the deposition rate was 1.0 μm/hr. The resistivity of the Ti-Al-N layer obtained by this method was 1×10^{-3} Ω · cm.

(4) A method of forming an n-type doped SiC layer on a metal substrate is described. The layer was formed by a high-frequency glow discharge decomposition method of 14.56 MHz, and the gas to be decomposed was a mixture of silane, methane (CH_4), and diborane (PH_3) in such a ratio that P/(Si + C) = 10 atomic %, the mixture having a pressure of 10 Torr. The substrate temperature was 300°C. At this time, by controlling the deposition time, the resultant n-type doped SiC layer had a thickness of 1,000 A. After the formation of the layer, a gold electrode was deposited on the SiC layer, and the resistivity of the SiC layer was measured and turned out to be 50 Ω · cm.

(5) A Pb layer having a thickness of 1 μm was formed on the surface of a stainless steel SUS 316 plate having a thickness of 1 mm by a vacuum deposition method. The layer was deposited in an Ar (99.9999%) atmosphere of 1×10^{-7} Torr at a substrate temperature of 200°C. Subsequently, a PbO layer was formed on the Pb deposited surface of the Pb deposited stainless steel plate by a sputtering method. The layer was formed in an Ar (99.9999%) atmosphere having an oxygen partial pressure of 2×10^{-4} Torr at a substrate temperature of 200°C, and the sputtering power was controlled such that the deposition rate was 3 μm/hr. The resultant sputtered layer was identified as PbO by structural analysis of X-ray diffraction. The resistiv-

ity of the PbO layer obtained by this method was 5 $\times 10^{-5}$ Ω·cm.

[0062] Also, in an example of using tin oxide, an In-doped tin oxide layer having a thickness of 0.5 μm was formed on the surface of a stainless steel SUS 316 plate having a thickness of 1 mm by a vacuum electron beam deposition method. The deposition was performed in an Ar gas atmosphere having a vaccum degree of 5×10^{-6} Torr at a substrate temperature of 300°C.

[0063] As described above, it has been confirmed that the current collector plate molded of the carbon molding material in which the surface-treated core metal plate is buried has better electric characteristics and smaller voltage loss than the current collector plate comprising the untreated metal plate.

Embodiment 7

[0064] This embodiment describes an example of integrally forming a current collector plate and an end plate for clamping the stack. The thickness of this current collector plate was increased in the stacking direction of the stack in order to keep the electric resistance extremely low. The current collector plate was made of a graphite plate having a thickness of 70 mm. The graphite plate had an electric resistivity of 1 mΩ·cm in the plane direction and 100 mΩ·cm in the thickness direction. Since the plate was as thick as 70 mm, a cell stack including the current collector plates was clamped by clamping members such as bolts, nuts and springs at a predetermined load. The clamping surface pressure applied to the electrode was checked with pressure-sensitive paper beforehand, and it was found that a surface pressure of 10 kgf/cm^2 was applied to the electrode and that the current collector plates had no cracking, remarkable distortion, or the like. At this time, a 0.5 mm thick tape made of polytetrafluoroethylene was wound around the bolt, and a 5 mm thick spacer made of polyphenylene sulfide was interposed between the current collector plate and the spring. In this way, a polymer electrolyte fuel cell stack was completed.

[0065] The current collector plates had a terminal section at one end, and a power output cable was connected to the terminal section. The specific structure of the terminal section is the same as that of Embodiment 1.

[0066] The stack was secured to an evaluation table, using a part of the clamping bolts, and an oxidant gas, a fuel gas and a coolant were supplied to the stack to generate electric power. As a result, it was confirmed that the stack could successfully generate a current up to a current density of 0.3 A/cm^2.

[0067] This embodiment has described the example of the current collector plate integrated with an end plate, but this current collector plate may be made to serve also as a separator by forming a gas flow channel on the anode or cathode facing side.

Industrial Applicability

[0068] The present invention provides a current collector plate composed mainly of a conductive carbon material which is low-cost, lightweight, and free from the fear of metal corrosion in manifold apertures. Particularly, by providing a terminal section in the vicinity of an inlet-side manifold aperture for a reaction gas, the electrodes are humidified by water generated by the reaction on the inlet side of the reaction gas supplied to the electrodes. As a result, the durability of the electrodes can be improved. Also, by forming a gas flow channel on one side of the current collector plate, the current collector plate can be integrated with the separator positioned at the end, so that the number of parts and the power generation loss due to the resistance components can be decreased. Accordingly, it is possible to reduce the cost of a polymer electrolyte fuel cell and improve the volume and weight efficiency thereof.

Claims

1. A polymer electrolyte fuel cell comprising: a cell stack of a plurality of unit cells, each of the unit cells comprising a hydrogen-ion conductive polymer electrolyte membrane, an anode and a cathode sandwiching said polymer electrolyte membrane, an anode-side separator having a gas flow channel for supplying a fuel gas to the anode, and a cathode-side separator having a gas flow channel for supplying an oxidant gas to the cathode; a pair of current collector plates that sandwiches said cell stack; and a pair of end plates that clamps said cell stack and said current collector plates under pressure, wherein said current collector plates comprise a conductive carbon material as a main component, and said current collector plates have a terminal section for connecting a power output cable in the vicinity of an inlet-side manifold for the fuel gas or the oxidant gas.

2. The polymer electrolyte fuel cell in accordance with claim 1, wherein said terminal section has a coating layer comprising a good electric conductor.

3. The polymer electrolyte fuel cell in accordance with claim 2, wherein the coating layer comprising a good electric conductor is provided in a region that extends from said terminal section to a part corresponding to an inlet-side part of the gas flow channel of the cell communicating with said inlet-side manifold.

4. The polymer electrolyte fuel cell in accordance with claim 3, wherein said coating layer comprises a metal plate.

5. The polymer electrolyte fuel cell in accordance with claim 4, wherein said metal plate is bonded to the current collector plates with a conductive adhesive.

6. The polymer electrolyte fuel cell in accordance with claim 1, wherein said current collector plates are such that the electric resistivity in the plane direction is smaller than the electric resistivity in the thickness direction.

7. The polymer electrolyte fuel cell in accordance with claim 6, wherein said current collector plates are such that the ratio of the electric resistivity in the plane direction to the electric resistivity in the thickness direction is 0.01 to 0.1.

8. The polymer electrolyte fuel cell in accordance with claim 1, wherein said current collector plates comprise a molded plate of a composite of the conductive carbon material and a binder.

9. The polymer electrolyte fuel cell in accordance with claim 8, wherein said molded plate includes therein a metal plate as a core member, and said metal plate, even at the manifold aperture of said molded plate, has no portion exposed to outside from said molded plate.

10. The polymer electrolyte fuel cell in accordance with claim 9, wherein said metal plate is coated with a conductive layer comprising noble metal, conductive inorganic oxide, conductive inorganic nitride, or conductive inorganic carbide.

11. The polymer electrolyte fuel cell in accordance with claim 1, wherein at least one of said current collector plates has a gas flow channel on an anode or cathode facing side and functions as an anode-side or cathode-side separator.

12. The polymer electrolyte fuel cell in accordance with claim 1, wherein said current collector plates are integrated with the end plates.

FIG. 1

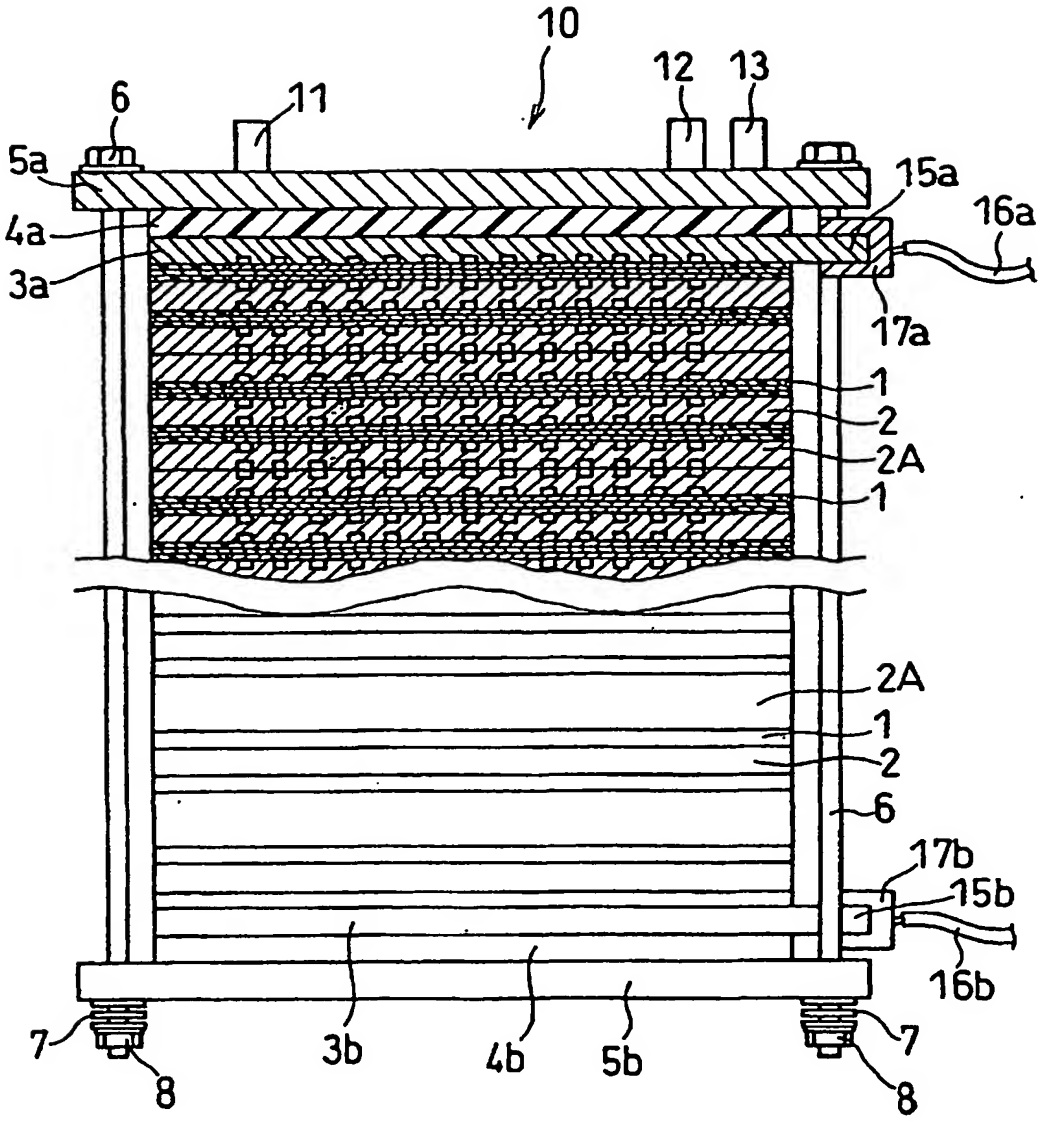

10
12
13
6
11
5a
15a
16a
4a
3a
17a
1
2
2A
1
2A
1
2
6
17b
15b
16b
7
3b
4b
5b
7
8
8

FIG. 2

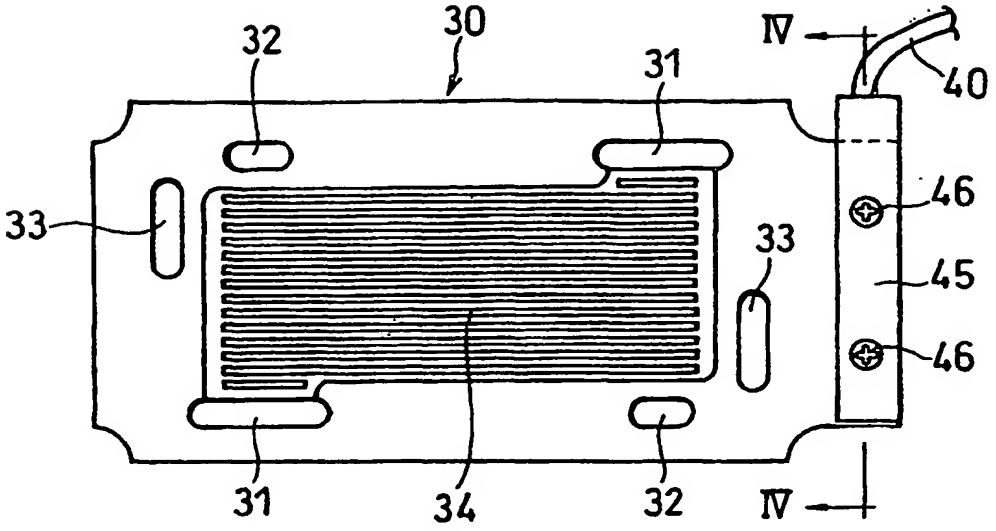

32
30
31
IV
40
33
46
33
45
46
31
34
32
IV

FIG. 3

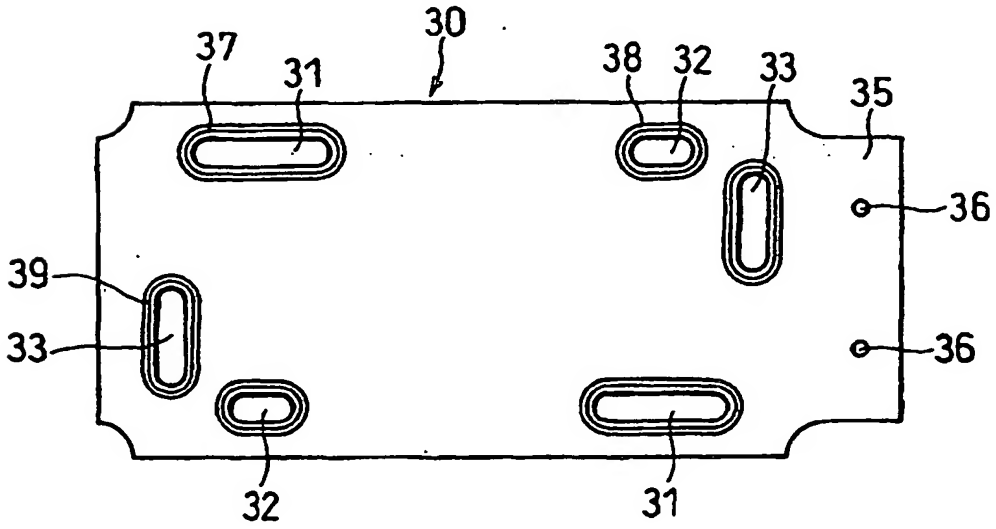

37
31
30
38
32
33
35
36
39
33
36
32
31

FIG. 4

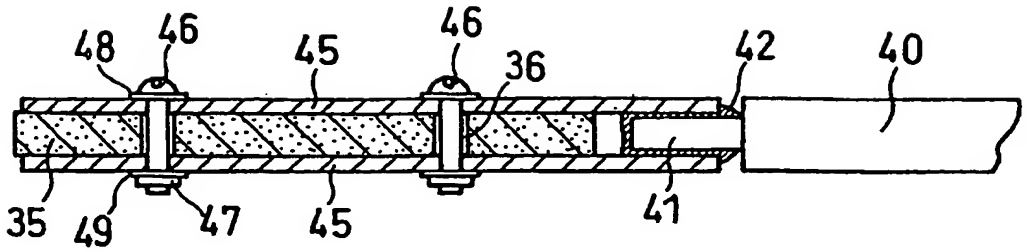


FIG. 5

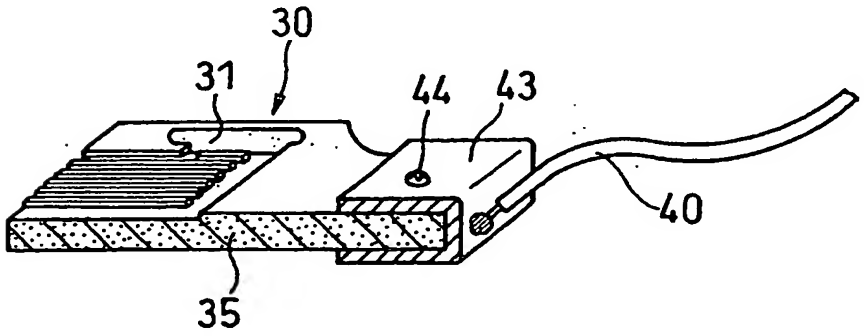


FIG. 6

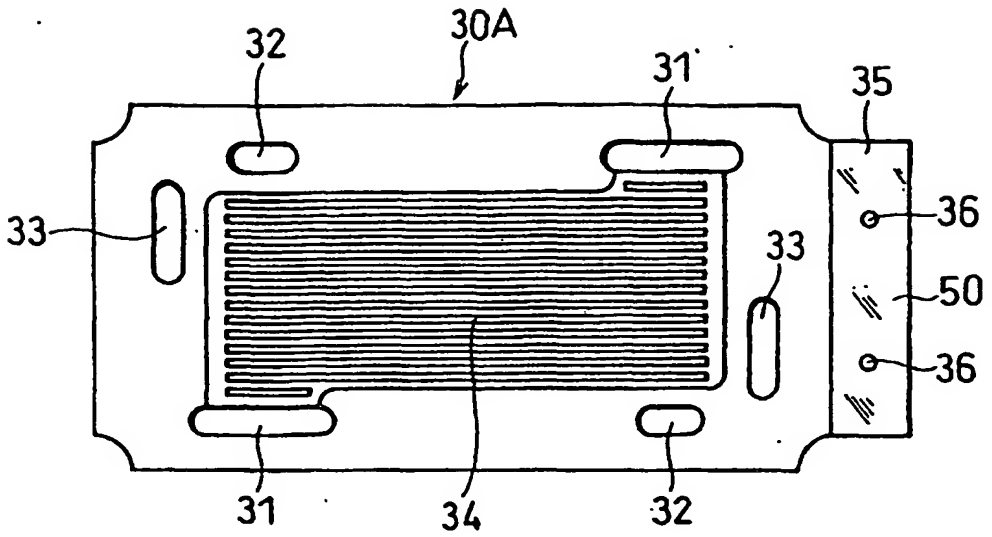

30A
32
31
35
33
36
33
50
36
31
34
32

FIG. 7

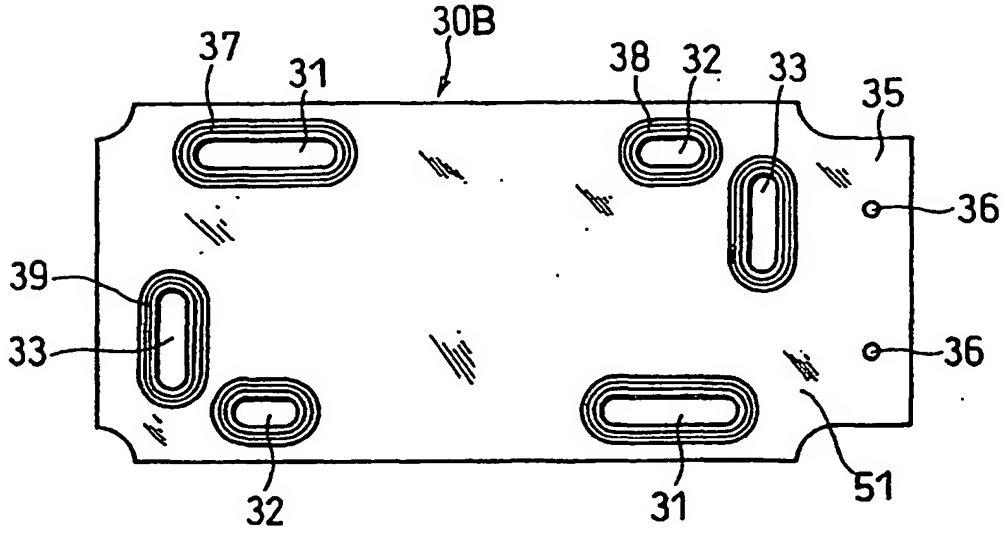

30B
37
31
38
32
33
35
36
39
33
36
32
31
51

FIG. 8

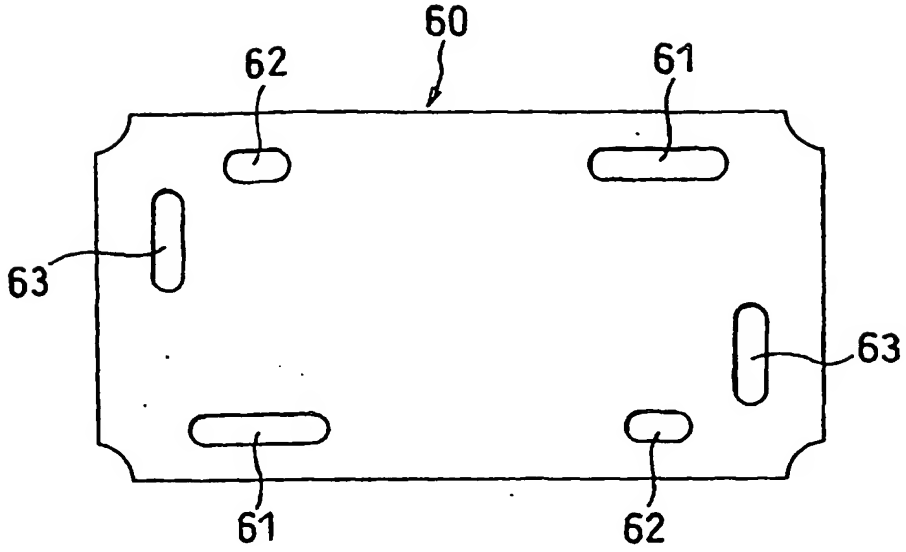


FIG. 9

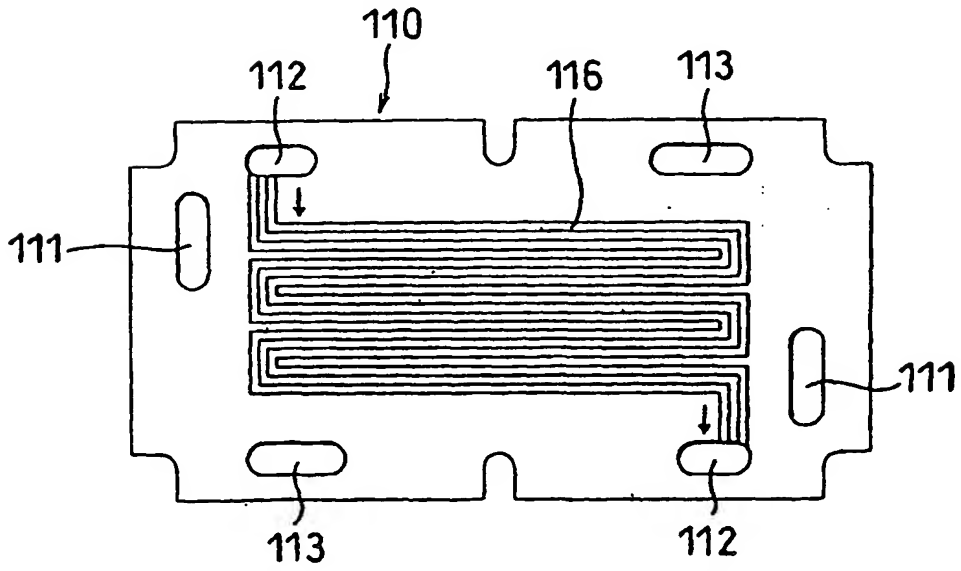


FIG. 10

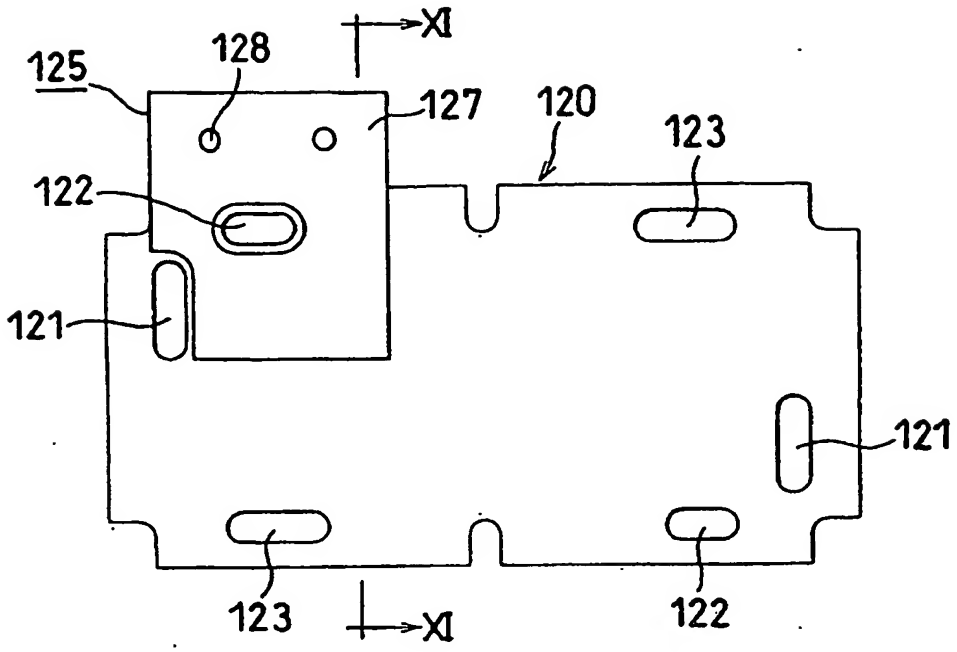


FIG. 11

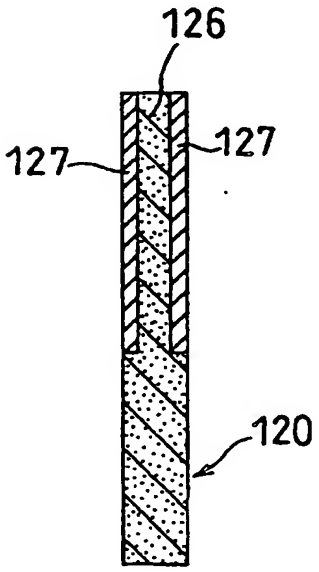


FIG. 12

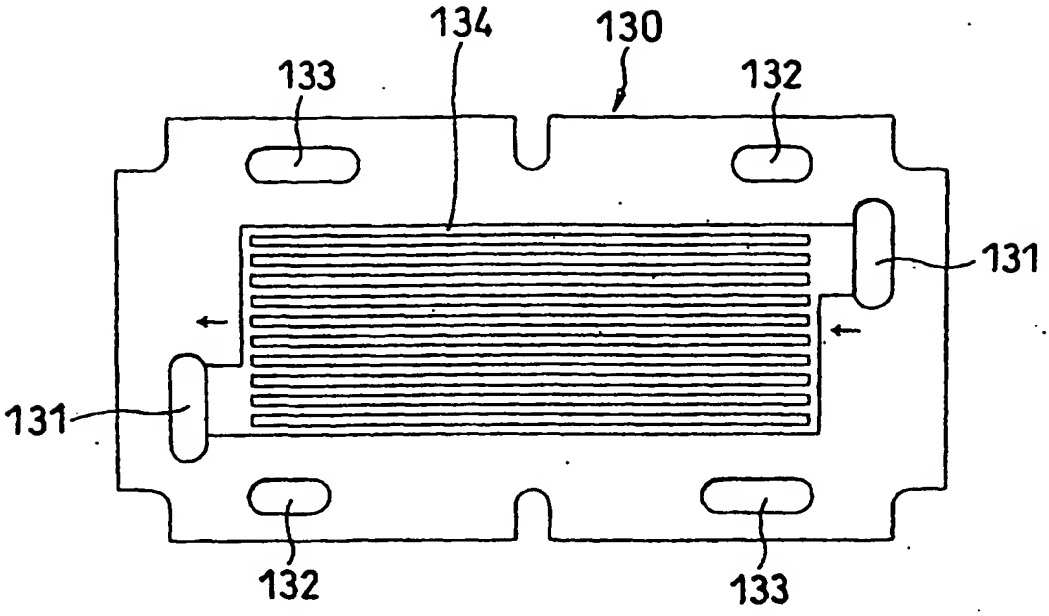

134
130
133
132
131
131
132
133

FIG. 13

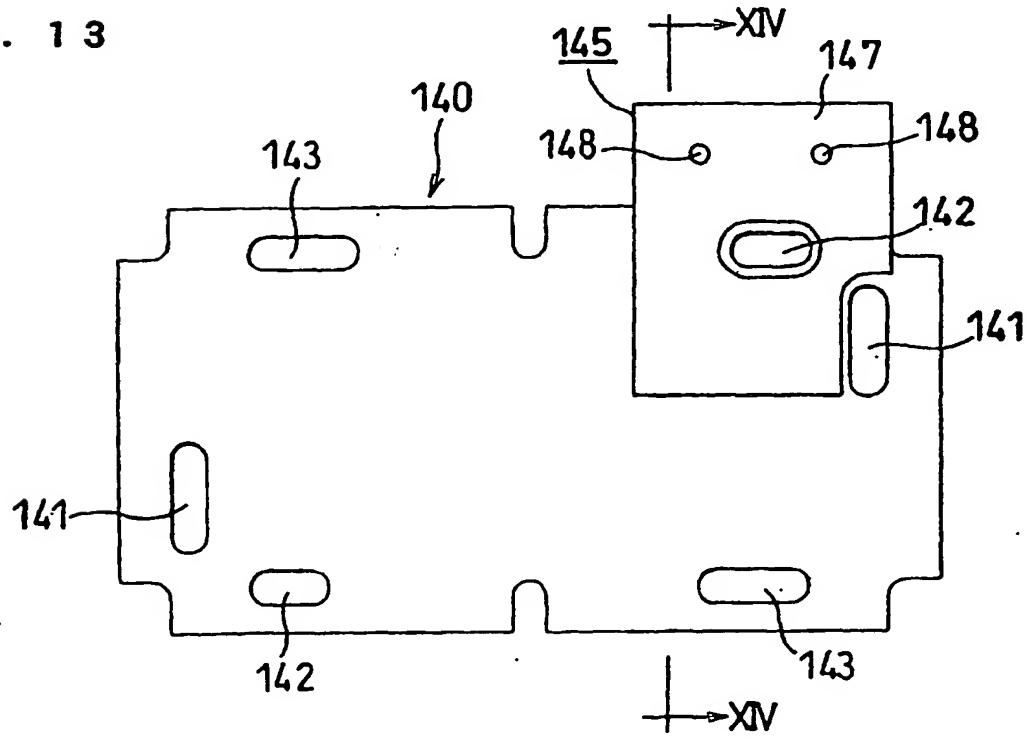

XIV
145
147
140
148
148
143
142
141
141
142
143
XIV

FIG. 14

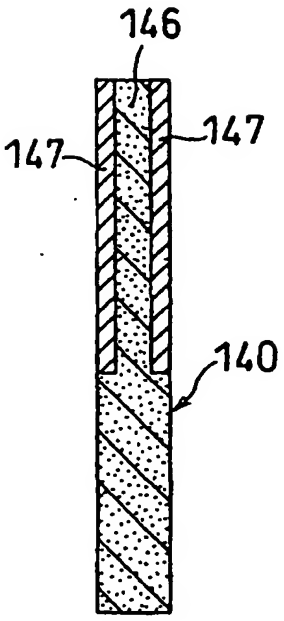


FIG. 15

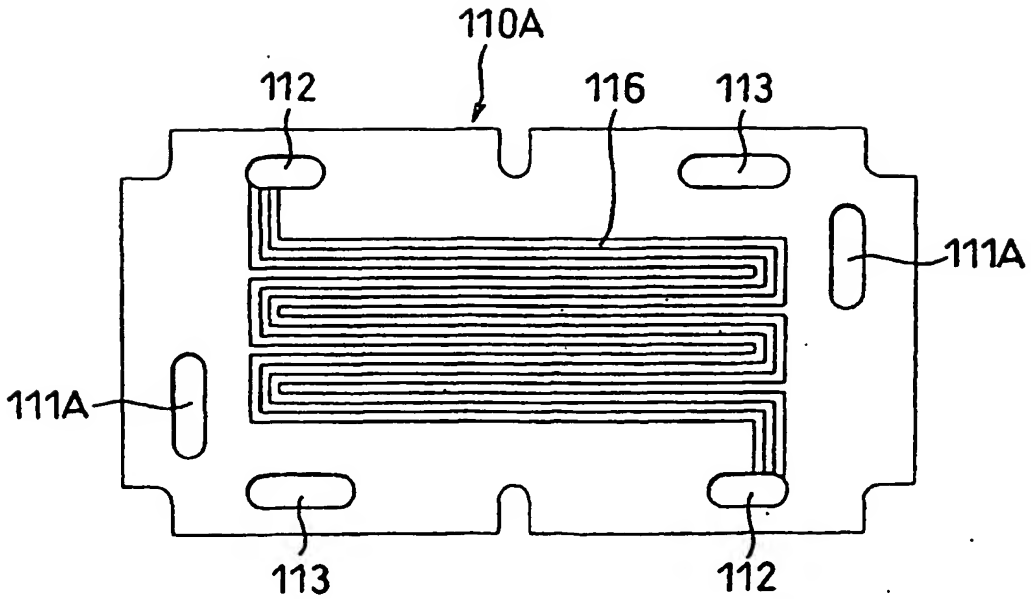


FIG. 16

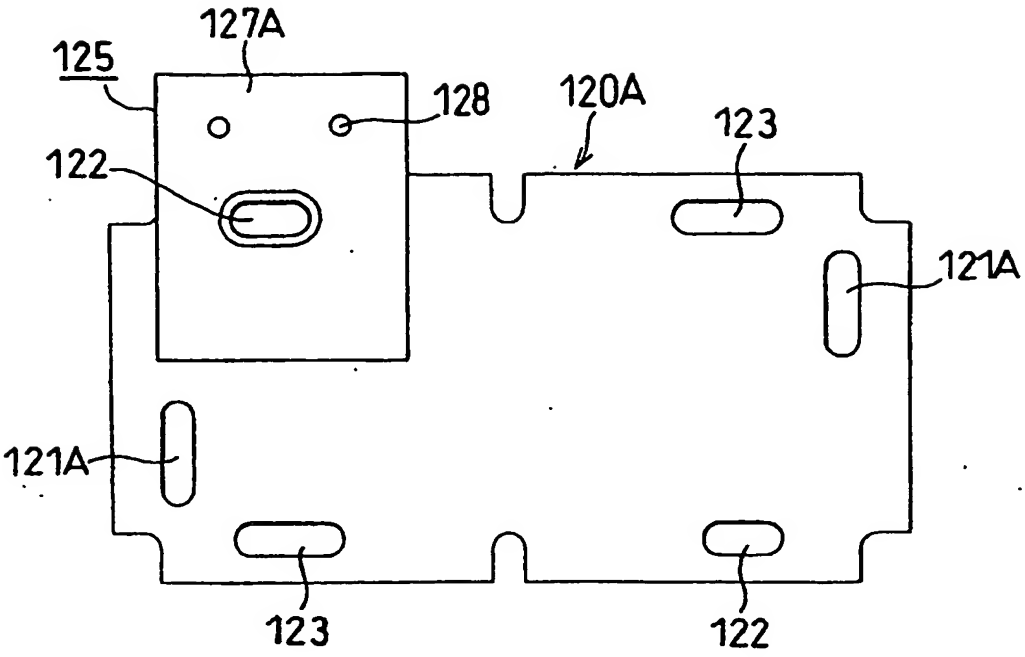

127A
125
128
120A
123
122
121A
121A
123
122

FIG. 17

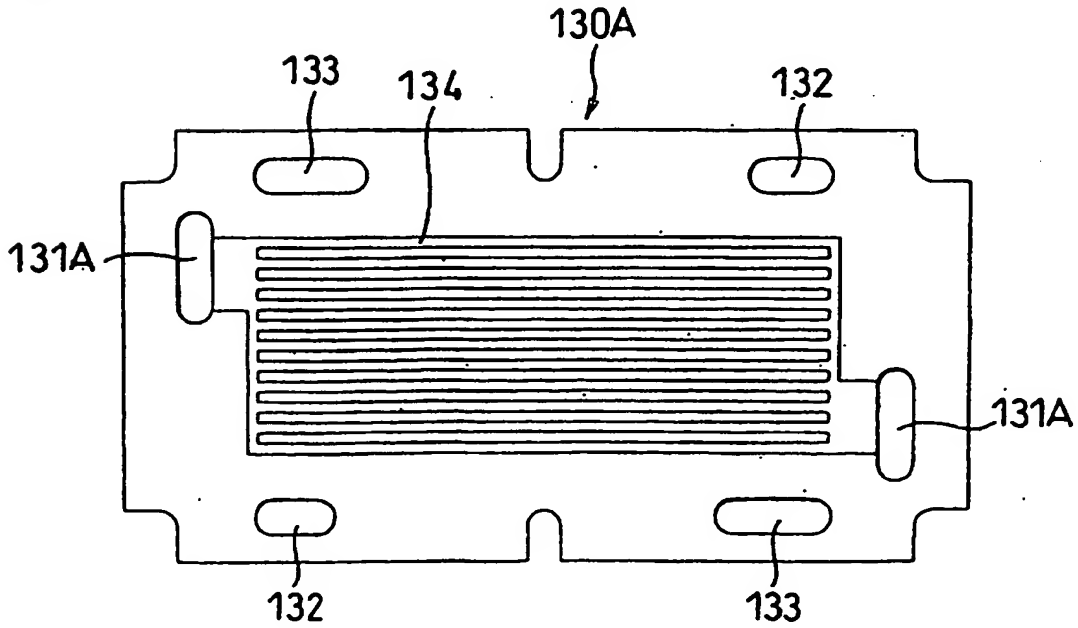

130A
133
134
132
131A
131A
132
133

FIG. 18

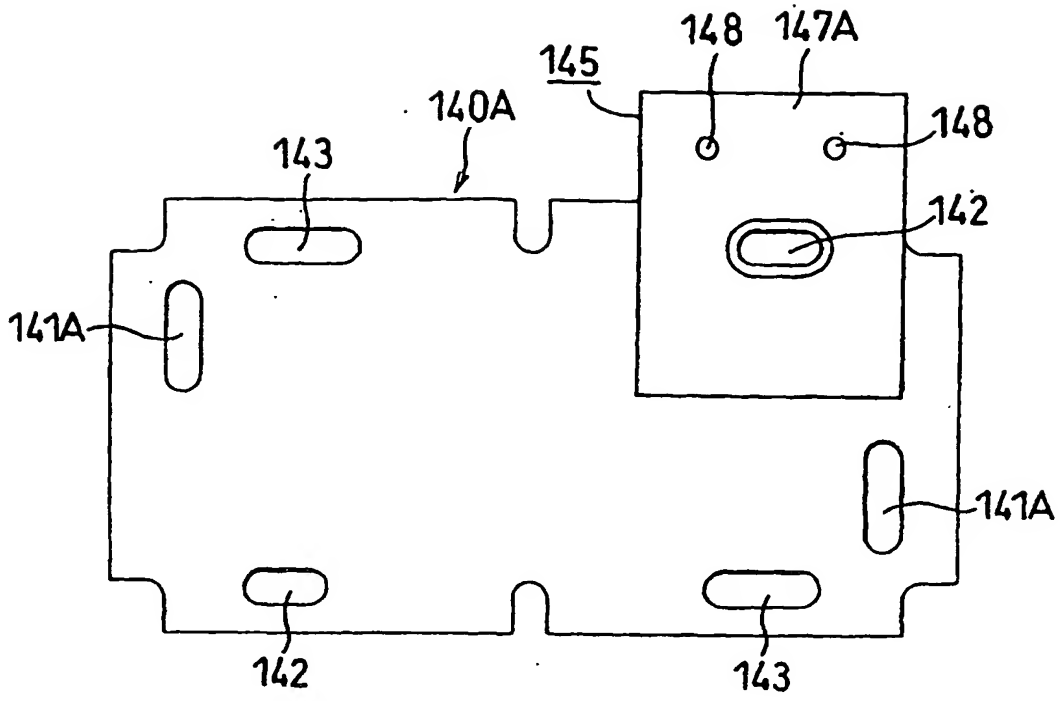

148
147A
145
140A
148
143
142
141A
141A
142
143

FIG. 19

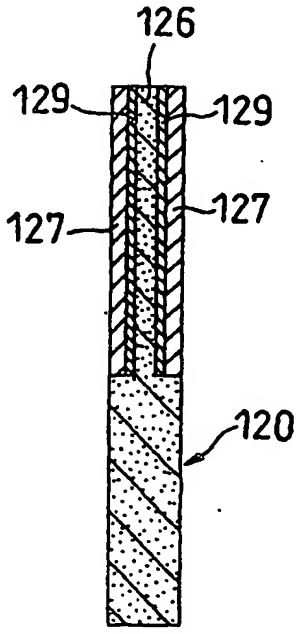

126
129
129
127
127
120

FIG. 20

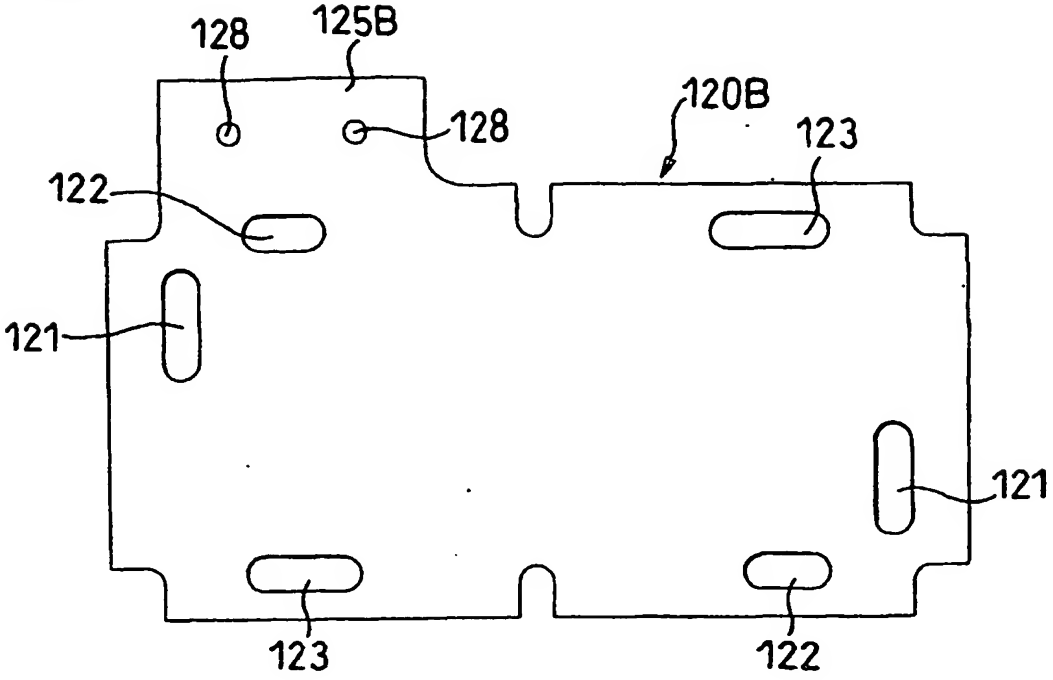

128
125B
128
120B
123
122
121
121
123
122

FIG. 21

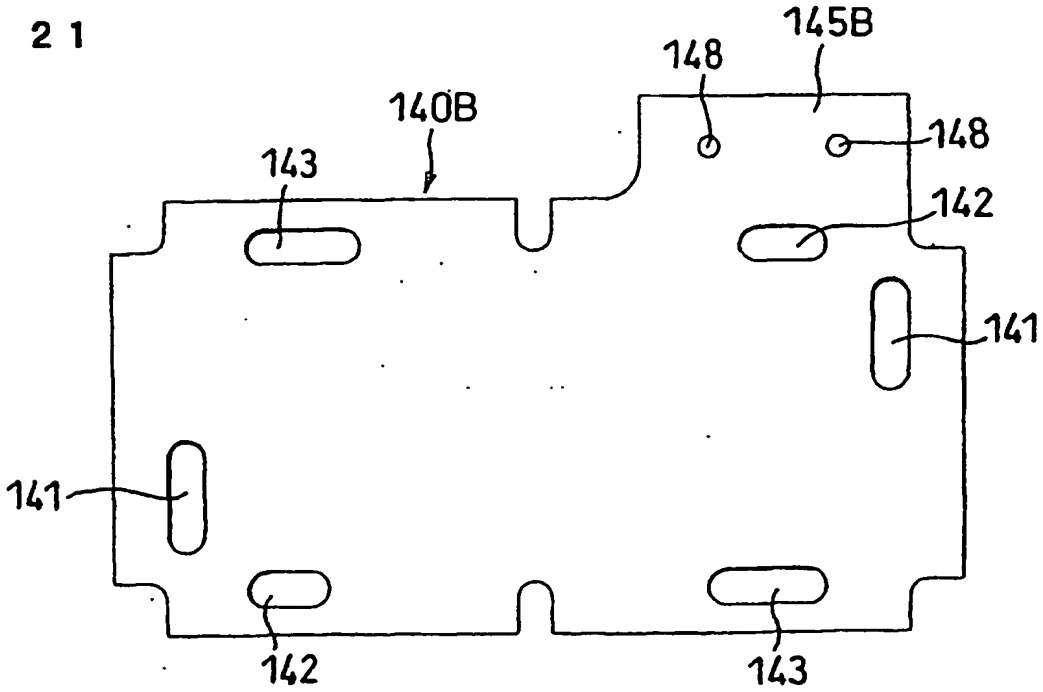

145B
148
140B
148
143
142
141
141
142
143

FIG. 22

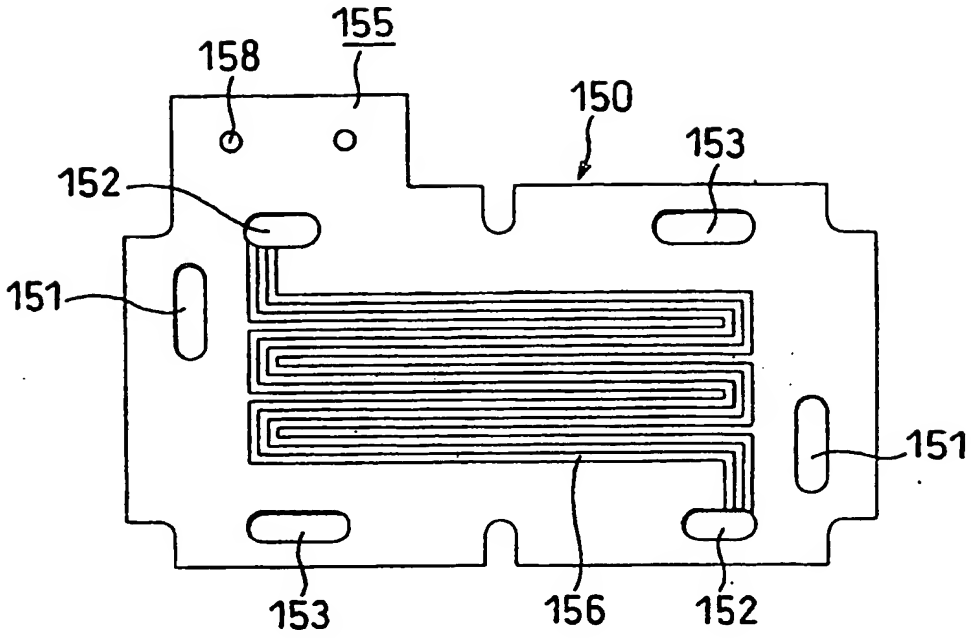

155
158
150
153
152
151
151
153
156
152

FIG. 23

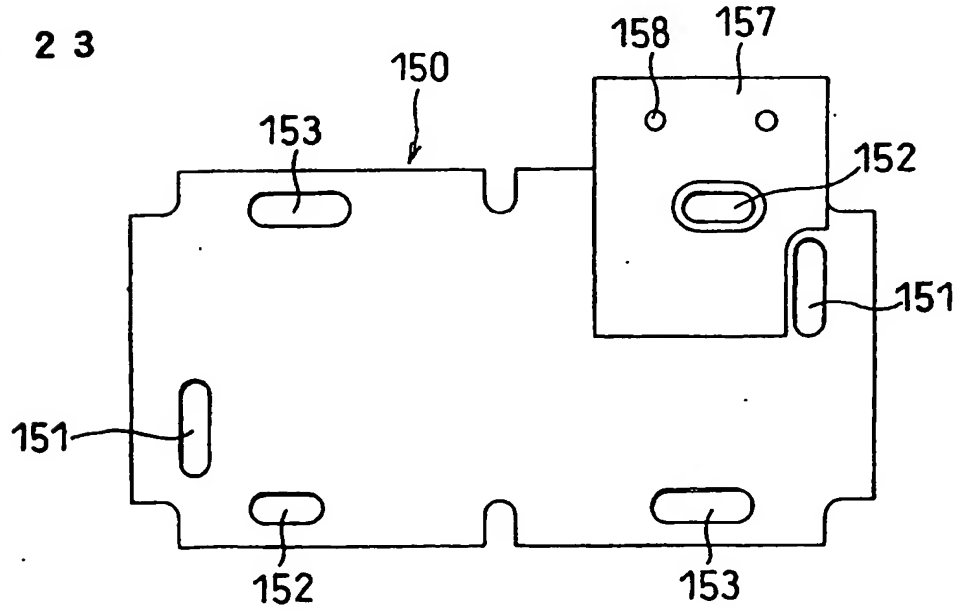

158
157
150
153
152
151
151
152
153

FIG. 24

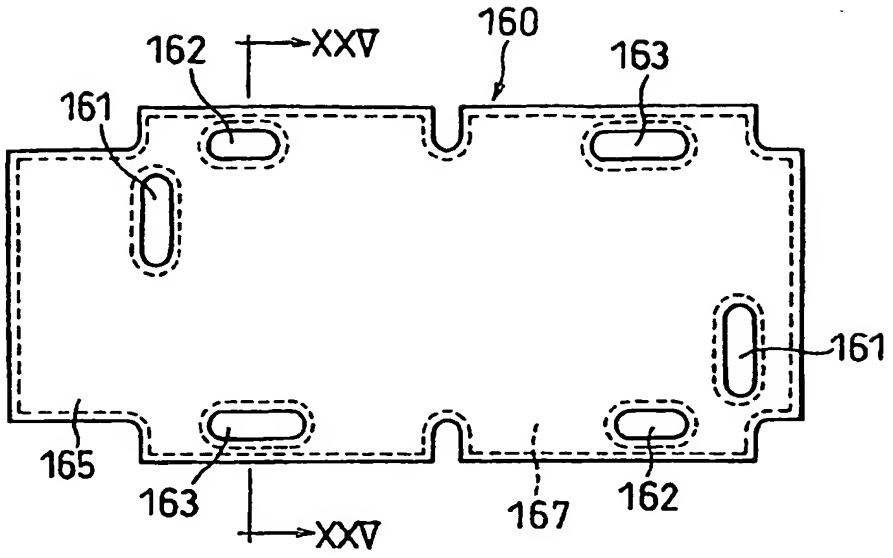


FIG. 25

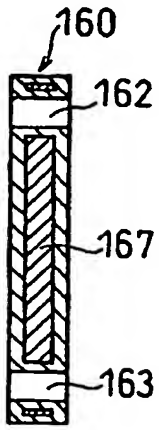
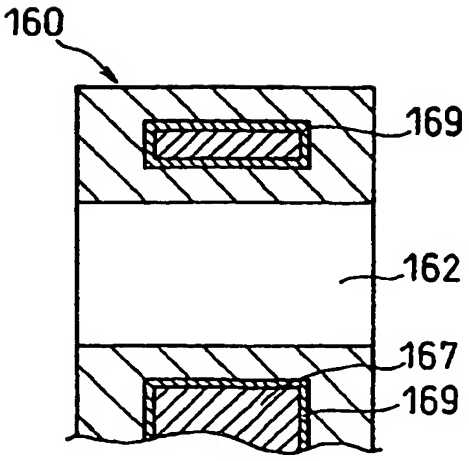


FIG. 26



INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP03/04723

A. CLASSIFICATION OF SUBJECT MATTER
Int.Cl[7] H01M8/02, H01M8/04, H01M8/10, H01M8/24

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
Int.Cl[7] H01M8/00-H01M8/24

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Jitsuyo Shinan Koho	1922-1996	Toroku Jitsuyo Shinan Koho	1994-2003
Kokai Jitsuyo Shinan Koho	1971-2003	Jitsuyo Shinan Toroku Koho	1996-2003

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y A	JP 58-166677 A (Mitsubishi Electric Corp.), 01 October, 1983 (01.10.83), Claims (Family: none)	1,2,8,11,12 3-7,9,10
Y A	JP 8-203553 A (Fuji Electric Co., Ltd.), 09 August, 1996 (09.08.96), Fig. 2 (Family: none)	1,2,8,11,12 3-7,9,10
Y A	JP 2001-176530 A (Toyota Motor Corp.), 29 June, 2001 (29.06.01), Fig. 3 (Family: none)	1,2,8,11,12 3-7,9,10

[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents:
"A" document defining the general state of the art which is not considered to be of particular relevance
"E" earlier document but published on or after the international filing date
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
"O" document referring to an oral disclosure, use, exhibition or other means
"P" document published prior to the international filing date but later than the priority date claimed
"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
03 July, 2003 (03.07.03)	15 July, 2003 (15.07.03)
Name and mailing address of the ISA/ Japanese Patent Office Facsimile No.	Authorized officer Telephone No.

Form PCT/ISA/210 (second sheet) (July 1998)

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP03/04723

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y A	JP 2001-43872 A (Aisin Seiki Co., Ltd.), 16 February, 2001 (16.02.01), Fig. 12 (Family: none)	1,2,8,11,12 3-7,9,10
P,A	JP 2003-45456 A (Hitachi, Ltd.), 14 February, 2003 (14.02.03), (Family: none)	1-12
P,A	JP 2003-100320 A (Toyota Motor Corp.), 04 April, 2003 (04.04.03), (Family: none)	1-12

Form PCT/ISA/210 (continuation of second sheet) (July 1998)